



RUH-284

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of: : C. Cooke  
HAGMEYER et al :  
Serial No.: 09/937,524 : Group: 1725  
Filed: September 25, 2001 :  
For: CATALYSTS...THEIR USE :

600 Third Avenue  
New York, N.Y. 10016

DECLARATION

Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

RECEIVED  
SEP 22 2003  
TC 1700

Sir:

Barbara Kimmich, familiar with the present application for Letters Patent hereby  
deposes and says as follows:

I am a graduate with a B.A. Degree from Franklin & Marshall College in  
Lancaster, PA in 1992 and a Ph.D Degree in Inorganic Chemistry from the University of  
Wisconsin-Madison, Madison, WI in 1998.

I have worked on numerous projects in the area of catalyst developments for  
organic reactions. I was employed at Brookhaven National Laboratory in Upton, N.Y.  
from 1998-2000. In 2000, I joined Celanese Chemicals. Since that time, I have been  
working on the development of improved catalysts for the manufacture of vinyl acetate  
from acetic acid, oxygen and ethylene.

During the impregnation of the sintered catalyst with the potassium acetate solution, it was observed, that the water adsorption of the catalyst had decreased by at least 50% and that the catalyst volume had decreased from ca.83 to ca. 47 ml.

Catalysts A and B were tested for the synthesis of vinyl acetate. For said testing, Catalyst B was diluted with P25 inert due to the extensive support shrinkage during the sintering step.

The performance of Catalysts A and B are shown in Table I

Table I- Vinyl acetate formation in the presence of Catalysts A and B, based on titania.

Catalyst Number	Catalyst A	Catalyst B
	Pd/Au acetates on P25 Pd/Au acetates on P25	
Support Size/shape	3.6 mm dia cylin	3.6 mm dia cylin
Reduction	1 h, 500°C 4% H <sub>2</sub>	1 h, 500°C, 4% H <sub>2</sub>
Sintering	none	800°C under 4% H <sub>2</sub>
Sel. to CO <sub>2</sub>	8.54%	16.4%
STY, g VA/1-hr.	793	672

CO<sub>2</sub>-selectivity in percent, based on reacted ethylene.

The following tests were conducted under my direction to demonstrate the superiority of the catalyst of the above application with respect to the catalyst of U.S. Patent No. 5,990,344.

#### Base Catalyst

A base catalyst was prepared by the procedure of Example 1 of the above application. Thus, P25 titania support (Degussa) was impregnated with palladium and gold acetate. After removal of the solvent, reduction using a mixture of 4 Vol. % H<sub>2</sub> in N<sub>2</sub> was conducted at a temperature of 500°C for one hour. The base catalyst was split into several samples, which were treated as follows:

#### Catalyst A

The base catalyst was impregnated with a solution of potassium acetate to yield a catalyst containing 40 g/l of potassium acetate. Catalyst A was prepared according to the instant application.

#### Catalyst B

The base catalyst was sintered at 800°C for 11 hours under a flow of a mixture containing 4 Vol. % H<sub>2</sub> in N<sub>2</sub>. According to column 2, lines 9-11 of Patent No. 5,990,344, the gaseous reducing agent was diluted with an inert gas such as nitrogen. After such sintering treatment, the catalyst was impregnated with a solution of potassium acetate.

### CONCLUSION

The above-mentioned data clearly demonstrates that the sintering treatment of U.S. Patent No. 5,990,344 has a detrimental effect on CO<sub>2</sub>-formation and the space-time yield if a catalyst is used, which is based on titania, produced through the flame hydrolysis of TiCl<sub>4</sub>. Hence, the above-mentioned data clearly reveal that the catalysts of the patent are different from the catalyst claimed in the above application.

The superior performance of the inventive catalysts in view of catalysts based on SiO<sub>2</sub> was already demonstrated in the specification. The comparison of Example 1 (P25 titania supported) with Comparison Example 5 (KA-160, Sudchemie, SiO<sub>2</sub>-support), as disclosed on page 24 of the specification clearly shows an improvement with regard to selectivity and space-time yield.

The undersigned declares further that all statements made herein of her own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the U.S. Code and that such willful false statements may jeopardize the validity of the above-identified application or any patent issuing thereon.

Celanese Ltd.

Date: Sept. 2, 2003

  
Barbara Kimmich

RECEIVED  
SEP 22 2003  
TC 1700